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SOLVABLE MODEL FOR OPTICAL AND RADIATIVE COLLISIONS,(U)
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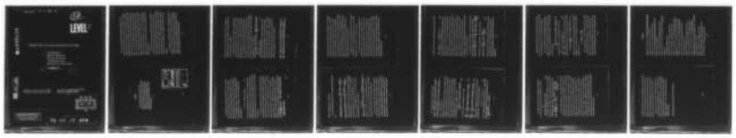

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Solvable Model for Optical and Radiative Collisions*

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Abstract

The related problems of optical and radiative collisions are examined theoretically. The actual R^{-n} potentials found in nature are simulated by powers of the hyperbolic secant, which enables one to obtain closed form solutions for the transition probabilities in the weak field regime.

Over the past several years, substantial attention has been given to the related problems of "optical" and "radiative" collisions (Cahuzac and Toschek 1978, Brechignac et al 1979, Falcone et al 1976a,b, Gallagher and Holstein 1977, Geltman 1976, Gudzenko and Yakovlenko 1972, Harris and Lidov 1974, 1975, Knight 1977, Lisitsa and Yakovlenko 1974, Payne et al 1977, Payne and Nayfeh 1976, Robinson 1979, Yeh and Berman 1979). In the optical case, foreign gas collisions enhance the probability of photoabsorption by an atom interacting with a radiation field detuned from resonance by more than the Doppler width. (For strong fields, the detuning is presumed to be in excess of the power-broadened homogeneous width.) Thus, without the collision, the transition probability is negligible.

A radiative collision is one in which excitation is transferred between two unlike atomic species due to the simultaneous effect of the collision between the atoms and their interaction with the radiation field. The photon is nearly resonant with the difference in excitation energy between the two atoms. Since the transition involves a change in state of both of the atoms, it is rigorously forbidden without the collision.

In this paper, we present solutions for a model version of these problems in the weak field limit. The models replace the true potentials that characterize the dynamics with powers of the hyperbolic secant. These retain the essential physics but enable one to obtain closed form expressions for transition probabilities.

The theoretical treatments are similar for the two problems. The relative nuclear motion is taken to be a classical straight line path, so that the atom-atom interaction generates a time-dependent potential for the electrons, whose motion is then described via the Schroedinger equation. Making the dipole approximation for the electron-field interaction, the electronic Hamiltonian for either problem is

$$H = H_a + V_c + e\vec{E}\cdot(\vec{r}_A + \vec{r}_B)\cos\Omega t, \qquad (1)$$

where H_a is the separated atom Hamiltonian, V_c the atom-atom interaction, $\vec{E}$ and the amplitude of the electric field (we assume linearly polarized light), $\vec{r}_A$ and $\vec{r}_B$ the sum of the coordinates of the electrons in each atom with respect to its respective nucleus, and Ω the frequency of the field.

For optical collisions, if one makes the further simplification that the effect of V_c may be adequately represented by its asymptotic van der Waals' R^{-6} energy, and that only two levels, the initial and final states of active atom A, designated $|1\rangle$ and $|2\rangle$, need be included in the expansion for the wave function, the Schroedinger equation becomes a pair of coupled equations for a two level system. Making the rotating wave approximation, the effective two level Hamiltonian is, in the "dressed atom" representation (Yeh and Berman)

$$H = \sum_{j=1}^{2}\left(\frac{\Delta C_{6j}}{2\nu R^6}\right)|j\rangle\langle j| + (\chi C_6/R^6)\left[|1\rangle\langle 2|\exp(i\nu t) + |2\rangle\langle 1|\exp(-i\nu t)\right], \qquad (2)$$

where C_{6j} is the van der Waals' constant of $|j\rangle$, $C_6 = C_{61} - C_{62}$, ν the Rabi frequency, χ the field strength in frequency units, and Δ the detuning from exact resonance.

One may approximately reduce the radiative collision problem to a two level system as well. Here states $|1\rangle$ and $|2\rangle$ are composites--in state $|1\rangle$ atom A is excited to A^* and B is in ground state B_o, while in $|2\rangle$, A is in ground state A_o and B excited to B^{**}. The transition proceeds via virtual intermediate states of the composite system. If one sums over these and assumes that the relevant frequency range is sufficiently small to treat the summations as essentially frequency independent, we obtain, after angle averaging, the effective two level Hamiltonian

$$H = \sum_{j=1}^{2}\left(\frac{C_{6j}}{R^6} + Q_j E^2\right)|j\rangle\langle j| + \frac{KE}{R^3}\left[|1\rangle\langle 2|\exp(i\Delta t) + |2\rangle\langle 1|\exp(-i\Delta t)\right], \qquad (3)$$

where we have again represented the atom-atom interaction by its leading term at large separations. Here C_{6j}, Q_j are the van der Waals and AC Stark coefficients for state $|j\rangle$, K is the indicated intermediate state factor, and Δ the detuning. The time dependent Schroedinger equation for state amplitudes a_1 and a_2 become, in an interaction representation, for optical collisions

$$i\dot{a}_1 = (A/R^6)\left\{\exp\left[i\left(C_6'\int_0^t dt' R^{-6}(t') + \nu t\right)\right]\right\}a_2, \qquad (4a)$$

$$i\dot{a}_2 = (A/R^6)\left\{\exp\left[-i\left(C_6'\int_0^t dt' R^{-6}(t') + \nu t\right)\right]\right\}a_1, \qquad (4b)$$

where $A = (X/v)\, C_6$ and $C_6' = (\Delta/2v)\, C_6$.

The corresponding equations for the radiative collision problem are

$$i\dot{a}_1 = a_2 (KE/R^3) \{ \exp [i (C_6 \int_0^t dt' R^{-6}(t') + \Delta_g t)] \}, \quad (5a)$$

$$i\dot{a}_2 = a_1 (KE/R^3) \{ \exp [-i (C_6 \int_0^t dt' R^{-6}(t') + \Delta_g t)] \}, \quad (5b)$$

where Δ_g is a "generalized" central detuning, defined to incorporate the AC Stark effect, i.e., $\Delta_g = \Delta + (Q_1 - Q_2)\, E^2$. These equations are to be solved subject to the initial conditions $a_1 = 1$, $a_2 = 0$ for $t \to -\infty$. In the weak field limit, $a_1 = 1$ for all t, and the transition amplitude, $a_2(+\infty)$, becomes

$$a_2(\infty) = -iF \int_{-\infty}^{\infty} R^{-3m}(t) \exp [-i (\lambda \int_0^t R^{-6}(t') dt' + st)] dt, \quad (6)$$

where $m = 1$, $\lambda = C_6$, $s = \Delta$, $F = K$ in the radiative problem, and $m = 2$, $\lambda = C_6'$, $s = v$, $F = A$, in optical collisions. (In this low power limit, the AC Stark effect is naturally neglected.)

Thus, for weak fields, the problem reduces to the evaluation of the integral in equation (6). For the actual R^{-n} potentials found in nature, exact analytic expressions for this integral are not known, although various approximations have been tried which do yield closed forms for radiative collisions. Geltman (1976) and Knight (1977) use a method that is equivalent to neglecting the

level-shifting term in the exponential of equation (6), and obtain a result proportional to the modified Bessel function K_1. Robinson extended their results by approximating the R^{-6} level shifting potential by a delta-function in time, obtaining, in addition to a term that behaves like the K_1 function, a term dependent on modified Bessel and Struve functions I_1 and L_{-1}. In the present work, we replace the R^{-3} and R^{-6} interactions by model potentials which closely resemble their time dependence but for which the integrals may be exactly evaluated in terms of special functions.

We begin with the case of radiative collisions. R^{-3} is replaced by $(\mathrm{sech}\,\pi t/T)/\rho^3$, R^{-6} by $(B/\rho^6)\,(\mathrm{sech}^2 \pi t/T)$, where ρ is the impact parameter, $T = 2\rho/v$, where v is the relative velocity.

B is chosen so that the phase induced by the model and true potentials are the same. The choice of parameters renders the model and true coupling potentials equal to one another at $t = 0$, and has the property that when $C_6 \to 0$ and $\Delta = 0$, the model and exact problems predict the same transition probabilities. (One could instead make the parameters a function of detuning, choosing them so that the transition probabilities with $C_6 = 0$ are the same at all frequencies. This proves to have a small effect.) The shape of curves (magnitude of potential versus time) for model and true potentials are very close.

For the model potentials, equation (6) becomes

$$ia_2(\infty) = \frac{K}{\rho^3}\int_{-\infty}^{\infty} \operatorname{sech}\frac{\pi t}{T} \exp\left[i(\mu \tanh\frac{\pi t}{T} + \Delta t)\right] dt$$
$$= (KT/\pi\rho^3)\int_{-\infty}^{\infty} \operatorname{sech} x \exp\left[i(\mu \tanh x + \beta x)\right] dx, \quad (7)$$

where $x = \pi t/T$ and $\beta \equiv \Delta T/\pi$.

The author was unable to locate this integral in the general tables available to him, although once he had succeeded in evaluating it, did find it in the more specialized literature (Buchholz 1969). There is some epistomological value in presenting the evaluation of the integral in equation (6), so we do so, rather than merely presenting the formula of Buchholz. We write

$$F = \int_{-\infty}^{\infty} \operatorname{sech} x \exp\left[i(\mu \tanh x + \beta x)\right] dx. \quad (8a)$$

If we differentiate F with respect to μ, we have

$$\frac{dF}{d\mu} = i\int_{-\infty}^{\infty} \operatorname{sech} x \tanh x \exp\left[i(\mu \tanh x + \beta x)\right] dx \quad (8b)$$

and

$$\frac{d^2F}{d\mu^2} = -\int_{-\infty}^{\infty} \operatorname{sech} x \tanh^2 x \exp\left[i(\mu \tanh x + \beta x)\right] dx. \quad (8c)$$

Performing parts integrations of equations (8b) and (8c), and rearranging terms, we obtain the differential equation

$$\mu F'' + F' + (\mu + B)F = 0 \ , \quad (9)$$

where ' denotes differentiation with respect to μ. This equation is to be solved subject to the boundary condition that at $\mu = 0$

$$F = \int_{-\infty}^{\infty} \operatorname{sech} x\, e^{i\beta x} dx = \pi \operatorname{sech}(\pi\beta/2).$$

If equation (9) is reduced to normal form, we have, with $\phi = \mu^{1/2}F$

$$\phi'' + \left[\frac{1}{4\mu^2} + 1 + \frac{\beta}{\mu}\right]\phi = 0, \quad (10)$$

which has the same form as the radial Schroedinger equation for a charged particle in a Coulomb field of coupling constant $-\beta/2$ and "angular momentum" $L = -1/2$. The lack of symmetry that prevails in the radiative collision problem according to the sign of the detuning may be inferred directly from the great difference between the wave functions in the Coulomb problem for attractive and repulsive potentials. If we consider the radiative collision problem where the final state has a van der Waals potential that is more attractive than that of the initial state, negative detuning corresponds to a repulsive Coulomb potential, and positive detuning to an attractive Coulomb potential.

With the given boundary condition, we thus find the transition amplitude proportional to a confluent hypergeometric function

$$ia_2(\infty) = \left(\frac{K\pi E}{\rho^2 v}\right) e^{-i\mu} \operatorname{sech}\frac{\pi\beta}{2}\, {}_1F_1\left(\frac{1}{2} + i\frac{\Delta\rho}{\pi v}, 1, \frac{3\pi C_6}{8\rho^5 v}\right). \quad (11)$$

This reduces at exact resonance to the Bessel function J_0,

$$ia_2(\infty) = \left(\frac{\pi K E}{\rho^2 v}\right) J_0\left(\frac{3\pi C_6}{16\rho^5 v}\right). \quad (12)$$

Equation (12) could be obtained directly by noting that the change of variable $\tanh x = \sin y$ casts equation (8a) into the form of a well-known representation of the Bessel function (Abramowitz and Stegun 1964).

The total cross section may be calculated by integrating the transition probability over impact parameter, yielding

$$Q = 2\pi \int_0^\infty |a_2(\infty)|^2 \rho \, d\rho. \tag{13}$$

Off resonance, this requires numerical integration or other approximate methods. The resonant case may be evaluated exactly, however (Gradshteyn and Ryzhik 1965). Thus, for zero detuning, we have

$$Q = \frac{2\pi k^2 E^2}{2^{3/5} v^{8/5}} \left[\frac{3\pi C_6}{16} \right]^{-2/5} \frac{\Gamma(\frac{3}{5})\Gamma(\frac{1}{5})}{\{\Gamma(\frac{4}{5})\}^3}. \tag{14}$$

For small impact parameters, μ becomes large, and one may use the asymptotic form of the confluent hypergeometric function. We find that the transition probability → 0 as ρ → 0, oscillating with increasing rapidity as ρ diminshes. These oscillations are a well-known characteristic of this problem (Harris and White). However, our observation that the envelope of the transition probability vanishes linearly near the origin is in disagreement with the extrapolated curve of Harris and White. Those authors present a curve whose envelope approaches a constant non-zero value as ρ decreases. This is hard to understand, since a steepest descent evaluation of the integral in equation (6), with the true potentials in the integrand, yields a result qualitatively similar to our own. (One does obtain a constant envelope factor at small impact parameter in the optical collision case.)

Sample numerical values of total cross sections are close to the "universal" curve of Payne et al (1977), who evaluated equations (6) and (13) numerically, in terms of dimensionless variables.

As we have indicated, an entirely analogous procedure can be applied to the optical collision case, where $\operatorname{sech}^2 \pi t/T$ replacing $R^{-6}(t)$ in both the exponential and the prefactor. For this problem, we wish to evaluate

$$F = \int_{-\infty}^{\infty} \operatorname{sech}^2 x \, \exp[i(\mu \tanh x + \beta x)] \, dx. \tag{15}$$

A differential equation in μ is again obtained, this time

$$\mu F'' + 2F' + (\mu + \beta)F = 0, \tag{16}$$

which, to within a trivial transformation, is the same as the confluent hypergeometric equation. The relevant solution this time is

$$F = \pi\beta \operatorname{csch} \frac{\pi\beta}{2} e^{-i\mu} {}_1F_1\left(1 + \frac{i\beta}{2}, 2, 2i\mu\right). \tag{17}$$

In normal form, this becomes a Coulomb radial equation for angular momentum L = 0. Equation (16) is solved subject to the boundary condition at μ = 0

$$F = \pi\beta \operatorname{csch} \frac{\pi\beta}{2}.$$

For the impact limit ν = 0 and the solution once more reduces to a Bessel function, this time $J_{1/2}(\mu)$. We note that this impact limit is exact, i.e., it is identical to the weak-field impact limit of the original R^{-6} problem. That the model gives the same result as the true potential in this case is not surprising, since

the impact limit assumes that "nothing happens" during the collision, i.e., that the result depends only upon the phase induced by the collision, not upon the shape in time of the potential function. Since the true and model potentials induce the same phase by construction, there can be no difference between their results.

In future work, we shall apply the model developed here to specific systems of experimental interest. We also note that this model may be solved analytically for fields of arbitrary strength. The solutions one obtains are not among those special functions familiar to physicists. We will report on this work also in a subsequent article.

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